Low-Frequency Dielectric Tests of PLA/Flax Sustainable and Degradable PCB Substrates

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Abstract— To pursue a move towards sustainability in electronics manufacturing, recently we presented a biodegradable printed circuit board (PCB) alternative substrate, where polylactic-acid (PLA) and flax-textile reinforcement forms a composite with flame retardancy. The novel PLA/Flax substrate is capable of being a core material for double-sided PCB designs. This paper investigates into the low-frequency (1 Hz - 500 kHz) dielectric capabilities of the substrates with Omicron Dirana dielectric spectrometer and Wayne-Kerr 6430A impedance analyzer. For the experimental, two generations of one-sided substrates were measured, where the top side was covered with 35 µm Cu layer with PLA foil and polypropylene (PP) foil (A and B groups consequentially) serving as bonding layer between the composite and the Cu. It was found that The PLA/Flax substrates have high permittivity values at low frequencies, while at high frequencies, the permittivity is lower than that of the most commonly used FR4. Moreover, the additional PP layer caused a further increase in the relative permittivity. The tan δ of PLA/PLA samples was higher over the investigated frequency range. This high value of relative permittivity and dielectric loss at low frequency is connected to interfacial polarization originated from the structure of PLA/Flax samples. A local peak of tan δ was observed at 500 Hz on PLA/Flax samples indicating the presence of orientation polarization.

Keywords— sustainable electronics, biodegradable PCBs, low-frequency dielectric tests, electric insulation

I. Introduction

Electronic devices are essential in everyday life, and demand is ever-increasing. Most electronic devices will become hazardous e-waste after a while, and the amount of this waste is projected to increase by 2 million tonnes annually [1]. A possible aspect of helping the electronics industry to turn to more sustainable solutions is to reduce the load on the environment and focus on novel, more environmentally friendly materials. Recently, it was shown that 70 wt% of the WPCB (waste printed circuit board) can be calculated from the glass fibre reinforced epoxy composites [2], so it is logical to aim at this value. This is a significant ratio in terms of PCBA (printed circuit board assembly) waste, and an aspect worth investigating and pursuing in advancing of the field.

Various experiments and materials were presented in the recent years ranging from green microelectronics [3] to devices realized with additive technologies on green substrates (e.g. paper) [4], however according to Soon [5], the terms of “green electronics”, “biodegradable electronic devices” can reach generally to two or three ranges higher than “biodegradable PCBs” from the aspect of literature keywords (quantitatively). The topic is still underrepresented, despite the research of different laboratories and research groups around the world. As for possible bio-sourced and degradable reinforcement materials, jute textile [6], hemp and sisal [7], banana fiber-based reinforcement [8], and flax were added to typically PLA (polylactic-acid) based resins, where PLA is a general occurrence as a resin in the composites

[9,10,11]. Recently, vitrimers were also applied in compositing sustainable PCBs [12], also compostable PVA and regular components were applied in an early demonstrator of a computer mouse [13].

As a lack in the findings, we can conclude, that most studies do not present flame retardancy and reinforcement, or the combination of these two. We improved upon our previous studies in the last decade by adding flame retardancy and bio-based reinforcement [14,15,16] to a novel substrate composite with the help of Meshining (Hungary). We focused on changing the classical FR4 (Flame retardant Class 4) substrate to polylactic acid (PLA)-based ones with natural (and enhanced) flax-fiber reinforcement. The performance of the new core is compatible with current subtractive PCB fabrication and classical surface mounting technologies (SMT). At the early stage of the research we were able to produce working Arduino Nano demonstrators [17], highlighting the competence and the complexity of the new substrate. An example of an assembled demonstrator is shown in Figure 1, where the smallest component pitch is ~0.5 mm.

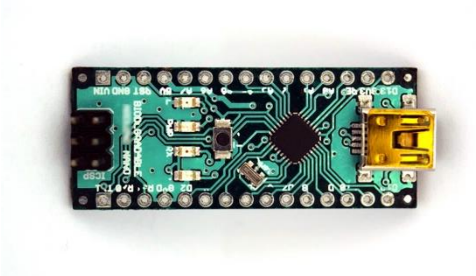

Fig. 1. Early demonstrator (Arduino Nano Rev 3.2 design on PLA/Flax circuits)

In this paper we do not aim for similar use cases, but focus on basic investigation of the substrate core. We present the results of low-frequency dielectric characterization of the substrate, and a general overview on the performance as a dielectric material. This is extending our knowledge on early high-frequency tests [18], which showed promising aspects of our environmentally friendly structure.

II. Experimental

A. *Boards (devices under test)*

The boards were prepared according to two groups. Both groups has the same core materials, with four layers of reinforcement and PLA as the resin in the composite structure – all handled with flame retardancy.

Group A is consisting of a one-sided, 1.55 mm thick board, which has a 35 µm Cu layer (HTE-ED-copper, electrodeposited, IPC-4562-compliant, from MSC Polymer, Germany) laminated on top with PLA foil acting as a bonding layer between the core and the conductor. Group B has additional polypropylene (PP) layer for improving adhesion with the same initial conditions.

The two board types (and the common cores) are the same like presented in the recent papers. [15,16,17] We have used FR4-type laminates (Flame Retard Type-4, 1.45 mm thick, as close to the PLA/Flax boards as possible) as references, with the same copper conductor applied to the surface.

B. *Experimental apparatus and methodology*

In the experimental methodology, we focused on measuring the dielectric parameters with low-frequency measurements (1 Hz...500 kHz) to reveal the electric insulation capability of the material. Below 5 kHz, the measurements were carried out by an Omicron Dirana dielectric spectrometer, while a Wayne-Kerr 6430A impedance analyzer was used at higher frequencies. The measurement voltages below and above 5 kHz were 100 V and 5 V, respectively. A guarded, circular electrode system with 10 cm^2 was applied. The equipment measured the capacitance and the tan δ values, and then the real part of permittivity values were calculated based on the size of the electrode system.

The ambiance during the measurements were kept at an air-conditioning controlled 20°C, which was supervised during the storage around the experiments and the measurement of the devices. The samples were stored in laboratory ambiance beforehand.

Figure 2 shows the setup for the measurements.

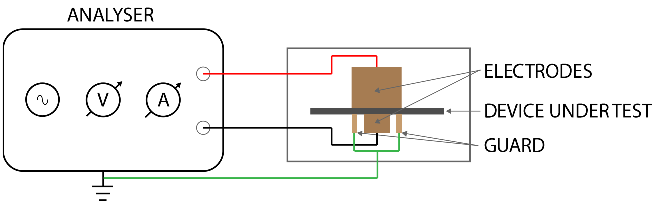

Fig. 2. Measurement setup with an analyser

III. Results and Discussion

The real part of permittivity measurement can be seen in Fig. 3. As Fig. 3 shows, both PLA samples (Groups A and B) have higher real part permittivities. According to dielectric theory, the real part of permittivity increases with a decrease in frequency. Both curves have a knee point of around 1 kHz. However, it is not seen very sharply. Nevertheless, the Group A sample curves have another knee below 10 Hz.

The real part permittivity of Group A samples above 500 Hz is lower than that of FR4 samples. The FR 4 samples' permittivity is around 5, at 500 kHz, while this parameter of Group A samples drops to around 2.5 which is almost the same value measured in [18]. The permittivity of Group B samples also decreases with frequency; however, their permittivity is higher than that of Group A samples in the whole frequency range. because this additional component can fill the cavities inside the Group B samples, increasing their permittivity. The value of the real part of the permittivity of Group B samples decreases to around 4.62 at 500 kHz. This value is also lower than the permittivity of the FR4 samples. Nevertheless, the aspect of the frequency dependence of permittivity in case of FR4 samples, is almost negligible in the whole frequency range.

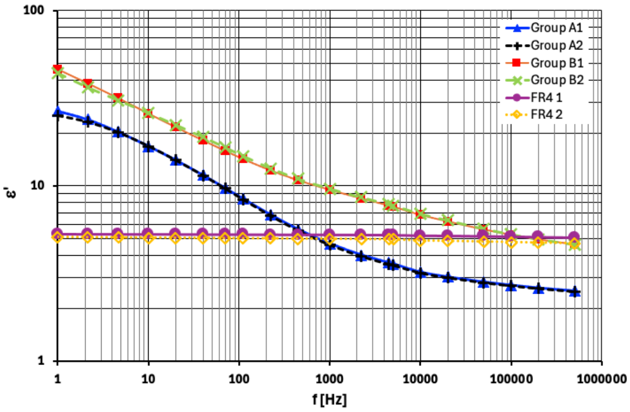

Fig. 3. The real part of permittivity vs. frequency.

The real permittivity values at some selected frequencies are summarised in Table I.

TABLE I. REAL PART OF PERMITTIVITIES AT SELECTED FREQUENCIES

freq.	Real part of permittivities of different samples					
	A 1	*A 2*	*B 1*	*B 2*	*FR4 1*	*FR4 2*
10 mHz	26.72	25.10	46.06	43.97	5.30	5.09
1 kHz	4.67	4.60	9.45	9.62	5.25	4.97
500 kHz	2.50	2.50	4.62	4.62	5.05	4.69

The PLA/Flax substrates have the highest permittivity values at low frequencies, while at high frequencies, the permittivity is lower than that of the most commonly used FR4. The material structure of the new biodegradable PLA substrate can explain this since samples were prepared using more components: two (Group A) or three (Group B) different materials were used. Hence, these samples can be considered a layered structure; therefore, it is suggested, that intensive Maxwell-Wagner (MW) polarization appears in them. The MW polarization is typical at low frequencies [19]. The low value of high-frequency permittivity can be due to the porous structure of the PLA samples, and cavities can exist inside the structure of samples, which will result in lower permittivity values.

Nevertheless, more investigation is required to reveal the root of this behaviour. The frequency dependence can be easily quantified by calculating the ratio of low- and high-frequency permittivities relative to a permittivity measured at a medium frequency. Table II. shows the ratios of 1 Hz and 500 kHz ε' values to 1 kHz permittivities. The ratio of ε' at 1 Hz to ε' at 500 kHz was also calculated to characterize the change of ε' in the whole frequency range. Table II data shows the real part of permittivity change in the whole frequency range, which is lower than 10% for FR4 samples, 5% for FR4 1 and 9% for FR4 2 samples. In contrast, this ratio is around 10 for Group A and B samples. Since the 500 kHz/1 kHz and 1 Hz/1 kHz epsilon ratios of Group A samples are more significant than those of Group B samples, it can be concluded that the frequency dependence of Group A samples is the highest also for lower and higher frequency ranges. Nevertheless, the high-frequency real permittivity value of Group B samples is almost twice that of the Group A sample because of the PP layer, as discussed above.

TABLE II. THE RATIONS OF REAL PART OF PERMITTIVITIES AT VARIOUS FREQUENCIES

freq. ratios	Ratio of ε' values at selected frequencies					
	A 1	*A 2*	*B 1*	*B 2*	*FR4 1*	*FR 4 2*
1 Hz /1kHz	5.72	5.46	4.87	4.57	1.01	1.02
500 kHz /1 kHz	0.54	0.54	0.49	0.48	0.96	0.94
1 Hz /500 kHz	10.67	10.05	9.98	9.51	1.05	1.09

Fig. 4 shows the results of tan δ measurement of samples. The results show that both groups of PLA samples have a greater tan delta than the FR 4 samples. Although the PLA samples' tan delta decreases with frequency, the FR4 samples' loss factor has a minimum value at 40 Hz; below and above this frequency, the tan δ increases. Nevertheless, the loss factor curve of FR 4 samples is flatter than the PLA samples' curves in the whole frequency range, hence the change of tan δ is significantly lower. Due to their layered structure, the loss factor of the PLA Group A and Group B samples is significantly higher than that of the FR4 samples tensive in the Hz range, which is a typical range of Maxwell-Wagner polarization. Therefore, the high low-frequency loss factor results from using different components (PLA, Flax and PP) for sample preparation.

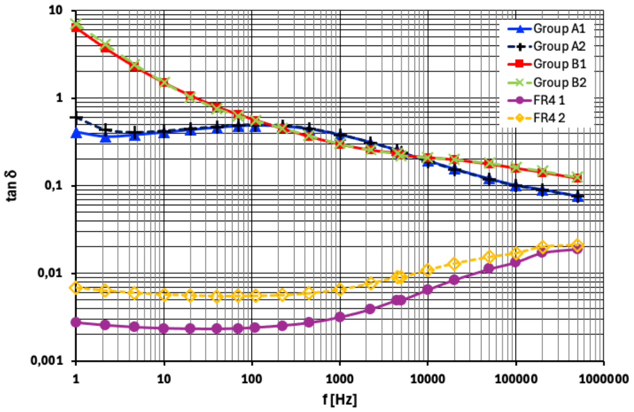

Fig. 4. Tan δ vs. frequency.

From the lowest frequencies, both PLA samples' tan delta gradually decreases, but a local peak can be observed in the case of Group A samples. This local tan δ peak at 500 Hz can be explained by the orientation polarization due to the polar molecules. The results show comparing to the convenient glass fibre reinforced epoxy composites the PLA composites has also higher dielectric constant and higher loss factor, especially at low frequencies, with strong frequency dependence.

Based on the results of dielectric measurements, the additional polypropylene layer slightly impairs the dielectric properties of the PLA substrate, namely increasing its relative permittivity and loss factor over a characteristic frequency range.

The current results are in alignment with higher frequency measurements presented in [15, 18].

IV. Conclusions

In this paper we have investigated two different groups of PLA/Flax composites serving as reinforced, flame-retarded substrates for PCB application, from the aspect of dielectric performance in the lower frequency regime. We have also used FR4-based substrates, as references.

The PLA/Flax substrates have high permittivity values at low frequencies, while at high frequencies, the permittivity is lower than that of the most commonly used FR4. Moreover, the additional PP layer caused a further increase in the relative permittivity. The tan δ of PLA/Flax samples was higher over the investigated frequency range. In the low frequency range, tan δ of the PLA/Flax samples was the highest. Moreover, the PP layer caused an additional increase of an order of magnitude in its value. This high value of relative permittivity and dielectric loss at low frequency is caused by interfacial polarization, which the structure of the PLA/Flax samples can explain.

A local peak of tan δ was observed at 500 Hz on PLA/Flax samples indicating the presence of orientation polarisation. This peak is overlapped by the extra loss caused by the PP layer on samples with an additional PP layer.

In the future, different substrates for bonding layer application between the copper and the core material has to be investigated, in order to find an appropriate optimum for the applied materials. Also, the values seem to limit the applicability of the bio-based, sustainable materials, however low-power applications, commercial, or electronics used in lower demand environments can still strongly benefit from the application of the bio-based substrate. New, impregnated and redesigned generations of materials are already in the measurement pipeline, so our future works will incorporate improvements upon the current results.

The characterization of similar substrates just started recently, and other more environmentally-friendly substrates, such as paper-based electronics will need in-depth analysis to have a thorough comparison between the future possible alternatives of FR4.

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REFERENCES

[1] Ruiz A, Latest Global E-Waste Statistics and What They Tell Us, 2023. october 16, Theroundup.org, accessed at 2024. 01. 27. Link: https://theroundup.org/global-e-waste-statistics/

[2] Khrustalev, D., Tirzhanov, A., Khrustaleva, A. et al. 2022. 2022. 2022. Sci Rep 12, 22199.

[3] B. Synkiewicz-Musialska et al., Preliminary characterization of glycerin, gelatin, propylene glycol, and cellulose based substrates for application in green microelectronics, September 2024, Scientiae Radices 3(3):212-217, 10.58332/scirad2024v3i3a04

[4] Ling, H., Chen, R., Huang, Q., Shen, F., Wang, Y., & Wang, X. (2020). Transparent, flexible and recyclable nanopaper-based touch sensors fabricated via inkjet-printing. Green Chemistry, 22(10), 3208–3215.

[5] Soon, C.F., Yee, S.K., Nordin, A.N. et al. Advancements in Biodegradable Printed Circuit Boards: Review of Material Properties, Fabrication Methods, Applications and Challenges. Int. J. Precis. Eng. Manuf. 25, 1925–1954 (2024).

[6] Larguech S., Triki A., Ramachandran M. et al. 2021. J Polym Environ, 29, 1240-1256.

[7] Kumar V., Gupta M. 2021. Comparative study of different natural fibre printed circuit board (PCB) composites. Materials Today: Proceedings, 44. 2097-2101.

[8] Bharath, K.N., Puttegowda, M., Yashas Gowda, T.G. *et al.* Development of banana fabric incorporated polymer composites for printed circuit board application. *Biomass Conv. Bioref.* 14, 12599–12612 (2024).

[9] R. Schramm, A. Reinhardt and J. Franke, "Capability of biopolymers in electronics manufacturing," *2012 35th International Spring Seminar on Electronics Technology*, Bad Aussee, Austria, 2012, pp. 345-349

[10] Giorgio Mattana, Danick Briand, Alexis Marette, Andrés Vásquez Quintero, Nico F. de Rooij, Polylactic acid as a biodegradable material for all-solution-processed organic electronic devices, Organic Electronics, Volume 17, 2015, Pages 77-86.

[11] C. Henning et al., "Realization of Double-Sided Wiring Boards of Biopolymer," 2020 43rd International Spring Seminar on Electronics Technology (ISSE), Demanovska Valley, Slovakia, 2020, pp. 1-6.

[12] Zhang, Z., Biswal, A.K., Nandi, A. *et al.* Recyclable vitrimer-based printed circuit boards for sustainable electronics. *Nat Sustain* 7, 616–627 (2024). https://doi.org/10.1038/s41893-024-01333-7

[13] Vicente Arroyos, Maria L K Viitaniemi, Nicholas Keehn, Vaidehi Oruganti, Winston Saunders, Karin Strauss, Vikram Iyer, and Bichlien H Nguyen. 2022. A Tale of Two Mice: Sustainable Electronics Design and Prototyping. In Extended Abstracts of the 2022 CHI Conference on Human Factors in Computing Systems (CHI EA '22). Association for Computing Machinery, New York, NY, USA, Article 263, 1–10.

[14] Csaba Farkas, László Gál, András Csiszár, Vincent Grennerat, Pierre-Olivier Jeannin, Pascal Xavier, Dániel Rigler, Olivér Krammer, Zbynek Plachy, Karel Dusek, Róbert Kovács, Anna Éva Fehér, Attila Géczy, Sustainable printed circuit board substrates based on flame-retarded PLA/flax composites to reduce environmental load of electronics: Quality, reliability, degradation and application tests, Sustainable Materials and Technologies, Volume 40, 2024, e00902

[15] V. Grennerat, P. Xavier, P. -O. Jeannin, N. Corrao and A. Géczy, "High-Speed Digital Electronics Board on a Novel Biobased and Biodegradable Substrate," *2023 46th International Spring Seminar on Electronics Technology (ISSE)*, Timisoara, Romania, 2023, pp. 1-5,

[16] A. Géczy et al., "Novel PLA/Flax Based Biodegradable Printed Circuit Boards," 2022 45th International Spring Seminar on Electronics Technology (ISSE), Vienna, Austria, 2022, pp. 1-6,

[17] A. Géczy et al 2024 Nanotechnology 35 435201

[18] V. Grennerat et al., "Exploring PLA/Flax Substrates for Antenna Applications: Assessing Moisture, Temperature and Dielectric Constant Homogeneity," 2024 18th European Conference on Antennas and Propagation (EuCAP), Glasgow, United Kingdom, 2024, pp. 1-5, doi: 10.23919/EuCAP60739.2024.10501367.

[19] A. K. Joscher, “Dielectric Relaxation of Solids,” Chelsea Dielectrics Press, 1983